Thermoluminescence of Roof Tiles Irradiated by Atomic Bombs in Hiroshima and Nagasaki

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Thermoluminescence of roof tiles irradiated by atomic bombs in Hirosima and Nagasaki has been observed for obtaining the distribution of gamma dose, hitherto unknown, from the bombs. The glow-curve from bomb radiation in the past was different due to the decay, in shape at the part of the low temperature, from that of the Co^{60} radiation at present. But nine samples out of thirty pieces of roof tiles showed that the shape of the glow-curve was similar to that of the artificial one at the part of the higher temperature. In case of eleven samples, the deformation of those glow-curves was observed at the part of the higher temperature and the rest of the samples did not show any thermoluminescence intensity. In addition, the thermoluminescence intensity of Hiroshima samples is about decuple as large as that of Nagasaki samples for the same dose.

INTRODUCTION

Not a few studies on measurements of the distribution of absorbed dose of atomic bomb radiation at Hiroshima and Nagasaki have been tried to make more quantitatively, the relationship between hazard and dose of radiation which has been discussed from the genetic and medical standpoint. Most of the radiation subjected from an atomic explosion appear in the form of fast neutron and Gamma-ray. About the estimation of neutron dose several results have already been reported[1]. However, those data have been unsatisfactory for the above purpose, because of social and technical difficulties when atomic bombs were dropped. No method has ever been found especially for estimating the gamma-ray component, except for indirect one through the value of thermal neutron dose.

Thermoluminescence has been used for radiation dosimetry and applied to age estimation in sediments. Thermoluminescence dosimeter has been developed by employing crystals of manganese-activated calcium fluoride or manganese-activated calcium sulfate[2,3]. This dosimeter has high sensitivity (doses as low as $20\mu r$), and excellent linearity to absorbed dose. On the other hand, thermoluminescence has been used to find the radiation dose absorbed by the natural substances in the past[4]. And measurements of the thermoluminescence of carbonate sediments[5] have been suggested several years ago to be successful in geologic age determination, and dating of ancient ceramic material by this

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method is now in progress[6]. Our experiment was carried out to apply this method to roof tiles of Hiroshima and Nagasaki[7]. The roof tiles were heated in manufacturing process and then exposed to the bomb radiation. The samples sustained, in fact, some information about the absorbed dose of the bomb radiation 18 years before, and thermoluminescence glow-curves could be recorded except for the samples exposed to the fire. When the sample is subjected to an artificial radiation of Co^{60}, its glow-peak height in the series of our experiment are proportional to various dose. Therefore the relative equivalent radiation dose can be acquired from the above curve by using the calibration curve. It appears that this method is effective for the direct measurement of the dose of bomb radiation. In this paper, some results are shown about the thermoluminescence of roof tiles irradiated by atomic bombs.

EXPERIMENTAL PROCEDURES

The apparatus used to measure the thermoluminescence consists of a light-tight furnance with a silver hot-plate and a roller for inserting the sample, a Toshiba 7696 photomultiplier (2″ϕ), a direct current amplifier and a two-pen recorder. The hot-plate on which the sample is spread, is equipped with a chromelalumel thermocouple, so that its temperature can be recorded simultaneously with the light intensity. The photomultiplier tube is insulated thermally from the hot-plate by holding filter, and connected optically through

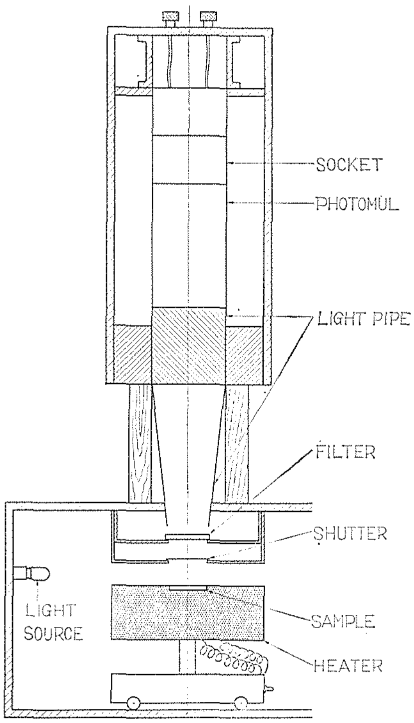

Fig. 1. Cross section of the furnance section and the light path.

the light pipe. The interrelation of the apparatus is shown in Fig. 1. For change in photomultiplier gain the light source is used to calibrate for it, and the shutter mechanism is prepared to keep the tube in the dark when the sample is inserted.

The roof tiles exposed by the bomb radiation were collected for samples, in Hiroshima and Nagasaki. Special care was taken in selecting samples to be exactly located and not to have been exposed to the fire occurred at the time of explosions.

The glow-curves resulting from the bomb radiation and the calibration curves were obtained by the following procedures. These samples were prepared by grinding a piece of roof-tiles with an agate mortar and sieving it (115 meshes) so that the standard grain sizes may be obtained. When the thermoluminescence curves from bomb radiation were needed, 300 mg of powdered samples was spread directly on the hotplate and pressed uniformly to the plate. The sample, when heated at the rate of about 75°C/min until the temperature reached 400°C, could record the glow-curve resulting from the bomb radiation. In the same manner, the background glow-curve from the thermal radiation was recorded. When the glow curves for gamma calibration were needed, the same sample was subjected to gamma radiation from Co^{60} (1000 curies) which could show an amount of absorbed dose, and likewise the new glow-curves were recorded to compare with the glow-curves from bomb radiation.

RESULTS

Thirty pieces of roof tiles in Hiroshima and Nagasaki were surveyed to observe the glow-curve resulting from bomb radiation. As the result, two thirds of the sample showed the thermoluminescence intensity except for the ones exposed to the fire, and nine pieces of them showed the shape of the glow-curves which we had expected. This type of the glow-curves obtained in this experiment is shown in Fig. 2 and Fig. 3. Where the glow-curve resulting from the Co^{60} irradiation is superimposed on the bomb glow-one for compari-

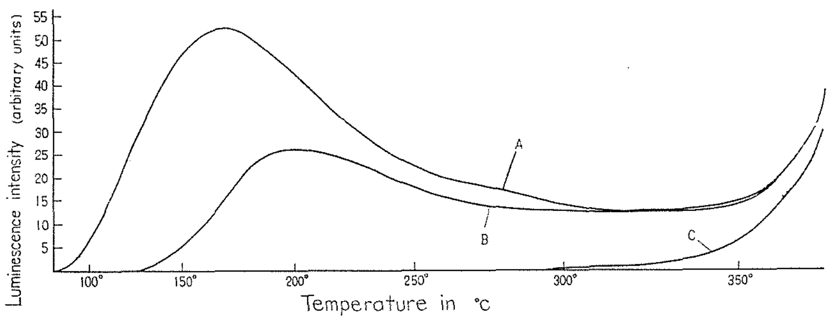

Fig. 2. Glow curves for roof tile at Hiroshima castle.
(A) T-L after irradiation with Co^{60} (Approx. 1×10^{3} r).
(B) T-L from bomb radiation.
(C) Black body radiation.

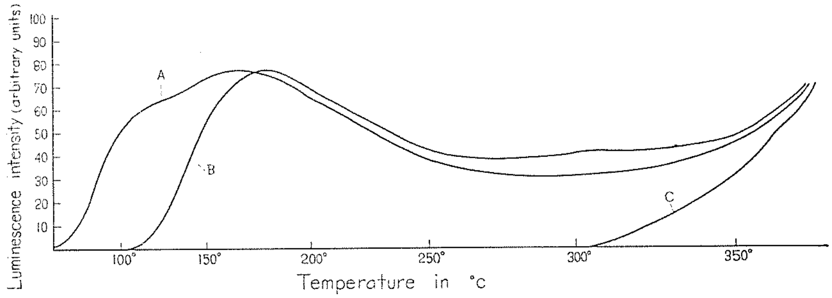

Fig. 3. Glow curves for roof tile at Nagasaki prison.
(A) T-L after irradiation with Co^{60} (Approx. 2×10^4 r).
(B) T-L from bomb radiation.
(C) Black body radiation.

son. The sample of Fig. 2 was collected at Hiroshima castle, 980 m from hypocenter of explosion, and the sample of Fig. 3 was collected at Nagasaki prison, about 250 m from the center. The value of the gamma dose of the Co^{60} irradiation adopted in this case is approximately the same as that of one irradiated by the bomb radiation. Ceramic material, such as roof tile, has many groups of trapping levels, so that its glow-curve dose not show any distinct glow peaks which are contributed by a few descrete trapping levels, but shows some very broad peaks.

A comparison of the glow-curves resulting from bomb radiation and the artificial glow-curves shows that the former glow-ones decay at the part of the low temperature (about 100°C). Such change of the glow-curves come from the decay at the normal temperature during the 18 years. As for the part of the high temperature, both of these glow-curves show nearly the same shape except for a little deformation which may be induced by grinding and annealling of the samples. Therefore, the samples of the above type are desirable for the measurements of the dose of bomb radiation.

On the other hand, the rest of the samples showed such glow-curves as in Fig. 4. In this case, it is seen at the part of the higher temperature that the thermoluminescence intensity increases gradually compared with the glow-curve

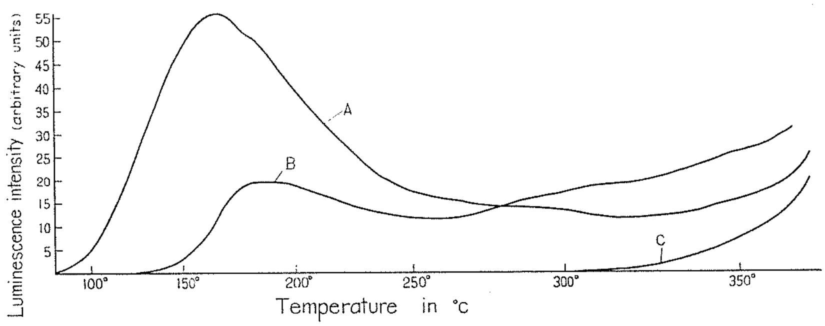

Fig. 4. Glow curves for roof tile at Hiroshima castle (Sample exposed to the fire).
(A) T-L after irradiation with Co^{60} (Approx. 1×10^3 r).
(B) T-L from bomb radiation.
(C) Black body radiation.

resulting from the Co^{60} irradiation. In our experiment the sampe annealed at 100°C for one week showed the same phenomenon in the glow-curves. Therefore this suggests that the above samples were exposed to the fire in low temperature and its glow-curves changed.

DISCUSSION

The thermoluminescence intensity shown by roof-tiles samples is contributed by natural radiation and bomb radiation. But the former is negligible compared with the latter, because the natural alpha radioactivity which was measured by means of an alpha scintillation counter gave the small value. The absorbed dose of the samples irradiated by bomb radiation is composed of four components; prompt gamma, fast neutrons, induced activity in the sample and fallout radioactivity. As for the samples treated in this experiment the contribution of the latter two components was small compared with the former two, because the samples exposed to the fire do not show any thermoluminescence intensity. In thermoluminescence dosimetry, the energy transferred to the sample by recoil of fast neutrons is about 1/20 as large as it is in the case of soft tissue because the atomic composition of the samle is much heavier than that of soft tissue. Hence it appears that the thermoluminescence intensity showed by the glow-curves is, for the most part, due to the gamma component of the bomb.

As is shown in Figs. 2 and 3, the thermoluminescence intensity at Hiroshima sample is about decuple as large as that of Nagasaki sample for the same dose. This difference is due to the mineral composition of the roof tiles samples. Roof tiles consist of various minerals and amorphous substances, some of which act the part of thermoluminescence phosphor. The samples separated into colourless mineral and coloured mineral by means of magnetic separator showed that the thermoluminescence sensitivity of the former was higher than that of the latter. In fact, it was observed in the microscopy, that Hiroshima samples contained much colourless minerals, such as quartz, and the sensitivity of them was so excellent that a bomb dose below 100γ could be measured.

CONCLUSION

As we have observed the thermoluminescence intensity of the samples gave some information about the absorbed dose of bomb radiation and most of it could be regarded as expressing the gamma component. Therefore, the thermoluminescence method may be an effective one for obtaining the distribution of gamma dose in Hiroshima and Nagasaki. Such problems remain yet to be solved as the experimental confirmation of a little deformation in the glow curves for the part of the high temperature (as is shown in Fig. 2), and of the exact separation of the contribution of the fast neutron dose.

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REFERENCES

(1) B. Arakatsu, K. Kimura, S. Shimizu, S. Kondo, F. Yamazaki, A. Sugimoto, *et al.*, "Reports on the Investigation of the Atomic Bomb Casulty", Vol. 1, Japan Society for the Promotion of Science (1953).

(2) J. H. Schulman, R. T. Ginther, R. D. Kirk and H. S. Goulart, *Nucleonics*, **18**, 92 (1960).

(3) B. Bjärngard, *Rev. Sci. Inster.*, **33**, 1129 (1962) ; AE-109 (Sweden), June, 1963.

(4) F. Daniels and D. F. Saunder, *ibid.* **111**, 462 (1950).

(5) E. J. Zeller, J. L. Wray and F. Daniels, *Bull. Am. Assoc. Petrol. Geologists*, **41**, 121 (1957).

(6) G. Kennedy, *Arch. News* **13**, 147 (1960) ; M. S. Tite and J. Waine, *Archaeometry* 4, 53 (1962) ; F. G. Houtermans, *Helv. Phys. Act.* 33, 595 (1961) ; Y. Ichikawa, *Journ. Nara Gakugei Univ.*, **11**, 55 (1963).

(7) T. Higashimura, Y. Ichikawa and T. Sidei, *Science*, **139**, 1284 (1963).